

Applicant : Vladimir Andreevich Bushuev
Serial No. : 09/649,431
Filed : August 25, 2000

and material being added in *italics*. Applicant states that no new matter has been added. A clean copy of the amended specification is attached as pages 1-12 of Exhibit B.

In the claims:

Please amend the claims as shown in page 14 of Exhibit A, which shows material being deleted in brackets and strikeouts, and material being added in *italics*. Applicant states that no new matter has been added. A clean copy of the amended claims is attached as page Exhibit B.

Remarks

Reconsideration and allowance of the present application in view of the foregoing amendments and accompanying remarks are respectfully requested.

In the Office Action dated November 29, 2001, the Examiner made final the restriction requirement, and withdrew from consideration claims 1, 2 and 5. Applicant has canceled these claims, without prejudice to present these claims in this or another application. In the Office Action, the Examiner stated that the Declaration and priority documents are missing from the file. According to applicant's records, a copy of the Declaration was filed with the application. Enclosed are the original Declaration as Exhibit C and the priority document as Exhibit D.

In the Office Action the Examiner stated that the title contains non-standard spellings for olefins and hydrocarbons. In response the title has been amended.

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In the Office Action, the Examiner rejected claim 4 as being allegedly indefinite. In response claim 4 has been canceled, without prejudice. Because this claim has been canceled, this rejection is now moot.

Claim 3 was rejected in the Office Action under 35 U.S.C. §102(b) by Dinulescu U.S. Patent No. 4,265,732. The Examiner stated that, with regard to claim 3, Dinulescu discloses a reactor for producing low-molecular weight olefins (column 5 line 42) by pyrolysis of hydrocarbons, comprising a housing (Figure 2a (5)) with directing stationary blades (4), an inlet nipple (Figure 2a) for supplying feedstock, and outlet nipple (figure 2a for carrying off cracked gas and a working wheel (1) provided with a blade crown (3), wherein the said housing has an annular cavity (Figure 2a) for circulation of hot pyrolyzed gas, said cavity containing the directing stationary blades and surrounding the blade crown of the working wheel along the periphery, and the said inlet nipple for supplying feedstock and outlet nipple for drawing off cracked gas communicating with the cavity (Figure 2a).

With regards to Claim 3, which the Examiner has rejected because it allegedly has no new features in respect of object described in Dinulescu's patent (US 4265732), applicant gives the further clarification.

The reactor shown in Fig. 2a of Dinulescu is a tandem combination of multi-stage axial compressor and axial turbine. Reactant flow entering through inlet nipple passes in series through cascades of stationary and rotating blades and then leaves reactor through outlet nipple. The design of this multi-stage axial compressor and axial turbine is of the conventional type (this is especially indicated in column 4, line 55 of the patent specification

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describing Fig. 2a), so it is impossible for the reactant flow or any part to move backwards from any intermediate point of the axial compressor and the axial turbine or from outlet of the axial turbine, to the inlet of the axial multi-stage compressor. Thus each particle of reactant flow passes through each cascade of stationary and rotating blades only once, and there is no possibility of repeated passing of reactant particles through the same cascades of stationary and rotating blades. Fig. 2b which shows monotonous rise of the temperature and pressure of reactant flow from inlet of axial compressor to border between the axial compressor and the axial turbine where the temperature and pressure reach their maximum (point B) and further so monotonous reducing of the temperature and pressure in the direction of axial turbine outlet.

In contrast, the reactor claimed by Applicant provides a cavity in the casing of the reactor which enables repeated passing of reactant particles through the same blade crown of rotor and stator. Such cavity is absent in the Dinulescu's reactor.

Applicant's claimed invention has the following advantages over Dinulescu's reactor:

- as a result of the said cavity being in new reactor "The period of time for heating the incoming feed/steam mixture from the temperature when entering into the reactor up to pyrolysis temperature is defined by duration of mixing with reacting mixture being processed and does not exceed 0.001 sec. It is negligible short time in comparison with the residence time of reacting mixture within a working space of the reactor" (see Application, page 8, paragraph 3 from the top). This overcomes the disadvantage in conventional cracking processes including process with the

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Dinulescu's reactor. The disadvantage consists in insufficient rate of feedstock heating - "because of this a starting amount of desired olefins formed at relatively low temperatures and carried further by feedstock flow through more and more intensively heated zones resides an excess time under conditions, when secondary reactions proceed with great intensity" (see Application, page 2, paragraph 2 from the bottom). So, in the new reactor a mixture of reactants is heated up "to maximum pyrolysis temperature practically instantly due mixing with hot pyrolyzed gases, thus resulting higher yields of low-molecular olefins" (see Application, page 6, paragraph 1 from the top);

- another advantage is that "the hydrodynamic regime realized within the reactor working cavity is close to regime peculiar to apparatus of ideal mixing, where concentrations of reactants are homogenized through-out the area of this cavity. The process essentially differs by this from process realized in traditional tubular reactors and from the process disclosed by US Patent 4265732 as well". Since the pyrolysis in the reactor working space proceeds in presence of just cracked products of high concentration, the pyrolysis reactions are accelerated due to autocatalysis phenomenon. Because of this the pyrolysis can be carried out also at lower temperature thus increasing the process selectivity (see Application, page 9, paragraphs 2 and 3 from the top);

- one more advantage is louver-damper configuration of the new reactor "enabling to make it in one-stage variant and of materials wide known and used presently in technique" (see Application, page 6, paragraph 1 from the top). This advantage is explained further as follows. Calculations demonstrate that for realization of the Dinulescu's reactor in one-stage variant (i.e. with using in axial compressor one rotating cascade of blades and one stationary

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cascade of blades) a peripheral velocity of blades should be about 2000 m/sec or higher. Such blades cannot be fabricated of presently known materials, furthermore these blades have to work at high temperatures. The energy needed for carrying out pyrolysis in the new reactor is transferred into reactant flow discretely, by relatively small portions, since "every particle of feedstock should pass over the rotor working blades several tens [of] times on average" (see Application, page 10, paragraph 3 from the top). This allows one to fabricate a new reactor in a one-stage variant applying materials presently known in technique, since the peripheral velocity of the blades can be 1000...4000 m/sec which is similar to the peripheral velocity of blades in modern gas-turbine engines.

Applicant believes that the Examiner may be interpreting the term "circulation" in the phrase "annular cavity for circulation of hot pyrolyzed gas" to have a different meaning than that intended by applicant. The word "circulation" could have two meanings. The first meaning is motion of a body on closed trajectory performing repeatedly as a rule. The second meaning is motion of a body on any curved trajectory occurring only once as a rule. The Examiner apparently interpreted the term "circulation" to have the second meaning and on this basis drew the conclusion about identity of features of the object described in Claim 3 and of the object shown in Fig. 2a of Dinulescu's patent, while applicant had in mind the first above mentioned sense of the term "circulation", which exactly corresponds with the Russian term "circulation" used in the PCT Application. In any case, claim 3 has been amended to eliminate any possibility of its wrong interpretation and now reads as follows:

A reactor for pyrolysis of hydrocarbons in production of low-molecular olefins comprising an inlet nipple for supplying feedstock, an outlet nipple for carrying off cracked gas, a working wheel with blades, stationary blades and a housing provided with a

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cavity, the said working wheel and the said stationary blades are located in the said cavity and the said cavity is configured so, that hot pyrolized gas being in it can repeatedly passes through the same blades of the said working wheel and through the same said stationary blades and the said inlet nipple and the said outlet nipple are communicated with the said cavity.

In view of the foregoing, applicant requests that the rejections be withdrawn and the application be allowed.

Applicant also encloses a Verified Statement Claiming Small Entity Status as Attachment E.


If a telephone interview would be of assistance in advancing prosecution of the subject application, the undersigned attorney invites the Examiner to telephone him at the telephone number provided below.

No fee is deemed necessary in connection with the filing of this Amendment. If any additional fee is necessary, authorization is hereby given to charge the amount of any such fee to Deposit Account No. 03-3125.

Respectfully submitted,

I hereby certify that this correspondence is being deposited this date with the U.S. Postal Service with sufficient postage as first class mail in an envelope addressed to:
Assistant Commissioner of Patents,
Washington, D.C. 20231.


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Process for producing low-molecular olefins[~~reactor for pyrolysis of hydrocarbons and apparatus for quenching cracked gas~~] *by pyrolysis of hydrocarbons and related apparatuses*

Field of the invention

15

The invention relates to the petrochemical industry[~~specifically~~]. *In one aspect, it relates to the process for production of low-molecular olefins by thermal cracking (pyrolysis) of hydrocarbons. In another aspect, it relates to the reactor for use in this process. In another aspect, it relates to apparatus for quenching cracked gas for use in this process.*

Background of the invention

20

At present a thermal pyrolysis of hydrocarbons is the basic process of commercial production of low-molecular olefins - ethylene and propylene. As a feedstock there are used hydrocarbons which molecule has two or more atoms of carbon. In industry there are generally used gases of petroleum refining, as well as naphtha and gas oil fractions.

25

In generally accepted technology a feedstock evaporated and mixed with steam, is supplied into a cracking tube located inside radiant section of pyrolysis furnace, wherein the mixture is rapidly heated. Pyrolysis reactions proceed with large absorption of heat. A cracked gas having the outlet temperature 750-950°C is quenched and transported into a gas fractionating plant, in which ethylene, propylene, butadiene, methane, hydrogen and other pyrolysis products are separated. Ethylene is the most valuable product of pyrolysis.

30

During pyrolysis of hydrocarbons a pyrocarbon is always evolved, part of which in the form of soot particles is carried away by flow of cracked gas, but the another part forms coke deposits on the walls both of cracking tubes and downstream apparatuses as well. Coke deposits increase a pressure drop through cracking tubes and deteriorate a proper heat transfer into reaction zone, resulting in overheating the cracking tubes, decrease in productivity of

pyrolysis plant and decrease in [~~low-molecular-olefins~~] yields of *low-molecular olefins*. Therefore coke deposits are periodically removed, usually this is performed by burning out with air or air-steam mixture.

Among reactions of thermal pyrolysis there can be distinguished: the primary reactions
 5 resulting in formation of olefins and the secondary ones, during which formed olefins are wasted. With temperature increase these reactions are accelerated, both primary and secondary, but the rate of primary reactions increases quicker than the rate of secondary ones. The rate of primary reactions does not depend on pressure, whereas the rate of secondary reactions decreases with pressure decrease. Therefore, to increase the olefin yields there strive to
 10 decrease hydrocarbon partial pressure in reaction zone and increase the process temperature within [~~regulation~~] *said* limits. The hydrocarbon partial pressure can be decreased by adding a steam-diluent. An optimal amount of steam-diluent depends on composition of hydrocarbon feed. For light feedstock - ethane or propane - the amount of steam usually is 20-40% of feedstock mass. For heavy feedstock, as gas oils, [~~a steam can amount to~~] *an amount of steam*
 15 *usually is* 80-100% of feedstock mass. It is undesirable to increase the pyrolysis temperature above 950-1000°C, because this accelerates sharply coke formation and causes growth of less valuable acetylene yield at the expense of ethylene.

Among disadvantages of commercial tubular cracking reactor the following is noteworthy:

20 a. it is necessary to transfer a large amount of heat into reaction zone through the cracking tube walls. Because of large heat flows the temperature of cracking tube wall much exceeds the temperature of a process stream causing an intensive coke formation and decrease in desired products yield. It is impossible to decrease the pressure in the zone of pyrolysis because of necessity to provide a high rate of feedstock flow through this zone required by
 25 conditions of heat transfer;

b. a rate of feedstock heating through cracking tube is insufficient. Because of this a starting amount of desired olefins formed at relatively low temperatures and carried further by feedstock flow through more and more intensively heated zones resides an excess time under conditions, when secondary reactions proceed with grate intensity. This disadvantage becomes
 30 apparent greatly in pyrolysis of wide petroleum fractions, such as naphtha or gas oil, which contain as high-molecular hydrocarbons cracked easily, as low-molecular hydrocarbons cracked at higher temperatures.

US Patent 5300216 discloses method and apparatus for thermal cracking hydrocarbons in presence of steam by passing through stationary shock wave of high intensity. A steam

superheated in tubular heater to the temperature about 1000°C is introduced at the pressure about 2.7 MPa through supersonic nozzle into reactor comprising series-positioned mixing and pyrolysis zones. In the mixing zone the hydrocarbon feed - ethane - preheated to the temperature about 627°C is introduced through mixers into supersonic flow of steam. Resulting mixture forms a supersonic process stream, which has a temperature lower than required to initiate pyrolysis reactions. Between the said mixing and pyrolysis zones a straight compression shock - continuous-standing shock wave - is created. ~~[When passing through]~~ In this compression shock a kinetic energy of the supersonic process stream is converted into the heat. Immediately downstream of the compression shock the velocity of the process stream falls to subsonic level, and the temperature rises up to about 1000°C at the pressure about 0.9 MPa abs. The process stream passes the pyrolysis zone for 0.005-0.05 sec. while its temperature decreases about to 863°C at the expense of heat absorbed by pyrolysis reactions. Conversion of ethane into ethylene achieves 70%. Cracked gas passes quenching apparatus and downstream heat exchangers, and further is transported to gas separation. In this apparatus all said above disadvantages of tubular pyrolysis reactors are eliminated. The feedstock reaches maximum pyrolysis temperature utmost rapidly, and the walls of pyrolysis section are not used for transfer of heat into reaction zone. But at the same time the required amount of steam per hydrocarbon mass ~~[rate]~~ must be about 500-667%. ~~[In connection with this]~~ *By reason of this* energy expenses per unit of produced ethylene are excessively high and unable to be essentially decreased. This renders the apparatus noncompetitive at current interrelation of energy costs with olefin costs.

US Patent 4426278 discloses tubular pyrolysis reactor including a steam superheater, an apparatus for mixing superheated steam with hydrocarbon and a cracking tube positioned inside radiation block. Coming from the superheater a steam with the temperature 1000-1500°C is mixed with hydrocarbon feed, so as to obtain immediately a temperature of resulting mixture required to initiate pyrolysis reactions. In this reactor the said above disadvantage (b) is partly eliminated. However, at the temperature 1100°C the steam must be supplied in amount 185-275% in a ratio to hydrocarbon mass ~~[rate]~~, and at the temperature 1430°C - about 120%. Preparation of superheated steam with such high temperature is extremely difficult and energy expenses are excessively high.

US Patent 3579601 discloses tubular pyrolysis reactor in which a feedstock is introduced into cracking tube at several points ~~[arranged]~~ *spaced out* along its length. Every portion of introduced feedstock reaches at once a temperature enough for starting pyrolysis reactions due to practically instant mixing with hot pyrolyzed gas incoming from upstream cracking tube run.

Pyrolysis is maintained further by heat supplied through cracking tube walls. This invention eliminates partly the disadvantage (b), but the disadvantage (a) remains. The yields of olefins and other unsaturated hydrocarbons are increased, and coke deposition and methane formation are decreased. The disadvantage of this invention is ~~[a-complicated]~~ *an intricate* configuration of the cracking tubes making difficulties for positioning it within radiant chamber of a heater.

USSR Author's Certificate 1189871 discloses process for thermal cracking naphtha and gas oils. A feedstock is separated into several fractions boiling out in intervals every 20-40°C. Such prepared narrow fractions are pyrolyzed in individual cracking tubes under optimal thermal conditions suitable for each fraction. This process enables to diminish the disadvantage (b), increase a yield of low-molecular olefins and decrease a coke formation, but technical realization of this process is associated with ~~[essential-complication]~~ *intricacy* of radiant chamber structure because it necessitates to create individual thermal conditions ~~[in]~~ *for* several cracking tubes under unsteady interrelation of feedstock flow rates in them.

US Patent 4265732 discloses process for thermal cracking gaseous hydrocarbon feed in a reactor constructed as multistage blading machine of axial type. A heat required for pyrolysis is generated directly inside a volume of reacting gas due to hydrodynamic drag of the rotor blades rotated therein by a drive. This invention eliminates completely the disadvantage (a), but the disadvantage (b) is not eliminated. Needed for process realization an axial type multistage blading apparatus capable of operation under the temperatures of hydrocarbon pyrolysis has not been fabricated.

US Patent 5271827 discloses tubular pyrolysis furnace provided with adiabatic tube reactor located between cracking tube outlet and an inlet of quenching apparatus. In the adiabatic tube reactor the pyrolysis is carried out at the expenses of intrinsic heat of reacting fluid without heat supply from outside. A use of the adiabatic tube reactor enables to economize on energy expenses required for pyrolysis.

Essential component of pyrolysis plants is the means for quenching cracked gas leaving reactor to the temperature of stopping undesirable secondary reactions. The quenching can be both direct - by injection of steam, water or light pyrolysis tar - and indirect - by using a heat exchanger. The direct quenching is usually applied in thermal cracking of gas oils. In thermal cracking of light hydrocarbons the indirect quenching in heat exchanger apparatus is usually applied generating simultaneously a high pressure steam.

PCT Application WO 95/32263 claims apparatus for quenching cracked gas. This apparatus comprises two spaces separated by a wall. A cracked gas flows in tubes, which form one of the said spaces, and a cooling water is boiled in the another space. Because of small

diameter of these tubes the apparatus of such type causes great flow resistance and is disposed also to coking. Usually the pressure drop through purged apparatus is not less 0.02-0.03 MPa, and in coked state it reaches 0.07 MPa and higher, that increases the pressure in upstream pyrolysis reactor and thereby decreases olefins yield. It could be possibly to decrease the pressure drop through apparatus by increase a diameter of heat transfer tubes, but such solution is impermissible since ~~[this]~~ it results in decrease of quenching rate.

Summary of the invention

The ~~[aim]~~ purpose of the invention is creation of a process for producing low-molecular olefins by thermal pyrolysis of hydrocarbons, reactor for ~~[pyrolysis of hydrocarbons and apparatus for quenching cracked gas, which serve for realization of]~~ use in this process and quenching apparatus for use in this process.

In this process:

- a. a heat is not transferred into reaction zone through walls closing this zone.
- b. a heating of hydrocarbon feed to pyrolysis temperature is performed for negligible time in comparison with duration of pyrolysis reactions.
- c. an amount of added steam-diluent per ~~[weight]~~ mass of hydrocarbon feed does not exceed the ~~[regulation]~~ limits accepted in existing pyrolysis plants.

~~[The first invention of the said above group is]~~ This purpose is reached in the process for producing low-molecular olefins by thermal pyrolysis of hydrocarbons~~[- This process comprises]~~ comprising ~~[the following stages]~~ preheating and evaporating a starting feedstock; mixing the same with a steam-diluent; heating resulting mixture to pyrolysis temperature in a blading rotary reactor ~~[by heat generated inside a volume of reacting mixture due to hydrodynamic drag of the rotor blades rotating therein]~~, quenching a cracked gas and subsequent separation of it, in this process the reacting mixture is heated to pyrolysis temperature ~~[for negligible time in comparison with a duration of pyrolysis reactions]~~ due mixing with hot pyrolyzed gas being circulated in a working cavity of the blading reactor.

Preferably preheating the feedstock and steam-diluent ~~[can be performed]~~ is performed in two stages, where the second stage is carried out in a heat exchanger by heat taken away from the cracked gas leaving the reactor.

In comparison with the process under US Patent 4265732 the present novel process increases desired olefins yield due to instant heating the reacting mixture up to maximum pyrolysis temperature by mixing with hot gases just pyrolyzed and yet circulated in working cavity of the reactor. The heat of cracked gas leaving the blading reactor is utilized for

preheating the feedstock and steam-diluent entering into reactor, that enables to simplify a structure of heat exchanger due to decrease in difference of pressure in heated and cooled sections and reduce also energy expenses for carrying out the process.

~~[The second invention of the group is the novel]~~ Purpose of the invention is reached also
 5 by using in the process for producing low-molecular olefins by thermal pyrolysis of hydrocarbons the reactor ~~[for pyrolysis of hydrocarbons. This reactor comprises]~~ comprising a housing ~~[provided]~~ with directing stationary blades, an inlet nipple for supplying feedstock, an outlet nipple for carrying off cracked gas and a working wheel provided with a blade crown. The housing has an annular cavity for circulation of hot pyrolyzed gas, where the directing
 10 stationary blades are located. This cavity surrounds the blade crown of the working wheel on periphery, and the inlet and outlet nipples are communicated with the said cavity.

~~[The housing of the reactor can consist of a casing and inner heat-resistant skin fastened together. The inner surfaces of the casing can be heat-insulated.]~~

As distinct from the reactor under US Patent 4265732 the ~~[present]~~ new reactor enables
 15 to heat the mixture of feedstock and steam-diluent to maximum pyrolysis temperature practically instant due mixing with hot pyrolyzed gases, thus resulting higher yields of low-molecular olefins. The another advantage of this reactor is louver-damper configuration enabling to make it in one-stage variant and of materials wide known and used presently in technique.

~~[The third invention of the group is the]~~ Purpose of the invention is reached also by using
 20 in the process for producing low-molecular olefins by thermal pyrolysis of hydrocarbons the apparatus for quenching cracked gas~~[-The apparatus comprises]~~ comprising a heat exchanger having two spaces separated by a wall~~[-for fluids being heated and cooled]~~. This apparatus is provided with a tee and with an ejector comprising a nozzle, admission and mixing chambers.
 25 The mixing chamber and one of the tee nipples are communicated with the space of cooled fluid, and the admission chamber is connected with the another nipple of the tee.

Such configuration of the apparatus for quenching cracked gas provides both a short time of quenching and small pressure drop through the apparatus as well, so that to obtain decrease in pressure in upstream reaction zone contributing to achievement of ~~[the main aim-]~~ increase
 30 in low-molecular olefins yield.

Brief description of the drawings

Fig. 1 is a schematic view of the installation for realization of the process for producing low-molecular olefins.

Fig. 2 is a sectional view of the reactor for use in the process for producing low-

molecular olefins by pyrolysis of hydrocarbons.

Fig. 3 is a cross-sectional view taken on line A-A in Fig. 2.

Fig. 4 is a sectional view of the radial working blade of the reactor.

Fig. 5 is a view on the arrow B in Fig. 4.

5 Fig. 6 is a sectional view of the apparatus for quenching cracked gas *in the process for producing low-molecular olefins by pyrolysis of hydrocarbons.*

Description of the preferred embodiments

10 The installation (Fig. 1) for realization of the process includes preheater 1, apparatuses 2 and 3 for quenching cracked gas, reactor 4, gas-turbine engine 5 connected with reactor 4 by shaft 6, and with the preheater 1 by exhaust pipe 7.

Preheating a feedstock and steam-diluent in the first stage is carried out in the preheater 1. The hydrocarbon feed from outside source (not shown in drawings) is conveyed by pressure into the preheater 1 configured as a shell-tube heat exchanger. The exhaust gas from the gas-turbine engine 5 is discharged into intertubular space of this heat exchanger. From outside
15 source (not shown in drawings) water is conveyed by pressure into the preheater 1 where the water is evaporated, and resulting steam-diluent is mixed with the hydrocarbon feed.

Preheating a feedstock and steam-diluent in the second stage is carried out in the apparatuses 2 and 3 for quenching cracked gas by utilization of heat of cracked gas leaving the reactor. Detailed description of the apparatus for quenching cracked gas will be given below.

20 The mixture of feedstock and steam-diluent from the apparatuses 2 and 3 for quenching cracked gas is conveyed into the blading reactor 4. Hot pyrolyzed gases are circulated ~~[along]~~ *within* the working annular cavity of the reactor, and the heat needed for pyrolysis is generated directly inside the volume of reacting mixture due to hydrodynamic drag effect of rotating working wheel provided with the blades. *Initial* heating ~~[the reaction mixture]~~ *of the particles*
25 *of feedstock up* to pyrolysis temperature is performed by mixing ~~[it]~~ *them* with the hot pyrolyzed gas for the negligible time in comparison with ~~[a]~~ duration of pyrolysis reactions. Detailed description of the reactor structure will be given below.

30 The cracked gas from the reactor 4 is conveyed into the quenching apparatuses 2 and 3 through interconnecting pipes having smooth shape to prevent formation of flow detachment zones. All these interconnecting pipes are equalized in volume.

The cracked gas is quenched in the apparatuses 2 and 3 and then transported into a gas fractionating plant (not shown in the drawings). In the Fig. 1 there are depicted two apparatuses for quenching cracked gas, but the number of this apparatus is not limited in the invention practical realization.

As a reactor drive the gas-turbine engine 5 is applied. Shown in Fig. 1 the gas-turbine engine is of a simple thermodynamic cycle without intermediate heaters and coolers of the working fluid. As a drive there can be used gas-turbine engines operating in more compound cycle, as well as steam turbine or electric motor.

- 5 The amount of [~~water~~] *steam* being mixed with hydrocarbon feed and tolerance final temperature depend on composition of feedstock. If a feedstock is the ordinary gaseous hydrocarbons, the amount of added [~~water~~] *steam* may be to 30-40% in ratio to hydrocarbon mass [~~weight-rate~~], and the temperature of reacting mixture after second preheating should not exceed 650°C. If the ordinary liquid hydrocarbons - such as naphtha or gas oils - are used as a
- 10 feedstock, the [~~water~~] *steam* may be added in amount to 80-100% in ratio to hydrocarbon mass [~~weight-rate~~], and relevant temperature of reacting mixture after second preheating should not exceed 550-600°C.

The basic parameters defining operation of the reactor are tied together by the following relationship:

15
$$\tau = V \times d \times H / P$$

where:

- τ (sec) is the mean residence time of reacting mixture in the reactor;
 V (m³) is the volume of the reactor working space;
 d (kg/m³) is the mean density of reacting mixture within the reactor working space;
 20 H (J/kg) is the energy transferred into reacting mixture within the reactor working space;
 P (W) is the power transferred into the reactor working space.

- The energy, which should be transferred into reacting mixture within the reactor working space, is near to amount of heat transferred into process stream flowing through a radiant coil in [~~traditional~~] *conventional* tubular pyrolysis furnaces operating with the same kind of
- 25 feedstock. In pyrolysis of ethane this energy should be about 2.5-3.4 MJ per kg of [~~steam deluted feedstock~~] *feed/steam mixture*. In pyrolysis of other kinds of hydrocarbon this energy should be about 1.7-2.3 MJ/kg.

The mean residence time of reacting mixture inside the reactor working space can be about 0.03-0.1 sec.

- 30 The period of time for heating the incoming feed/steam mixture from the temperature when entering into the reactor up to pyrolysis temperature is defined by duration of mixing with reacting mixture being processed and does not exceed 0.001 sec. It is negligible short time in comparison with the residence time of reacting mixture within a working space of the

reactor.

The mean density of reacting mixture is defined by average pressure, by average temperature in working space of the reactor and by average molecular ~~[weight]~~ mass of reacting mixture.

- 5 The average pressure within working space of the reactor can be arranged of 0.05-0.2 MPa (abs), preferably 0.08-0.12 MPa (abs).

10 The average gas temperature in the reactor working cavity depends on the feedstock composition, assigned conversion and residence time within reaction zone. Light feedstock and high conversion require the higher temperatures, but heavy feedstock - as atmospheric and vacuum gas oils - and low conversion require lower temperatures.

15 Operator sets the gas temperature in working cavity of the reactor and the rates of hydrocarbon feed and water for preparing steam-diluent. The set temperature is maintained by automatic control system regulating a rate of fuel gas feeding the gas-turbine engine. So the temperature in the reaction zone of the reactor is regulated as in principle as it is done in the ~~[traditional]~~ conventional tubular furnace - by changing a rate of fuel gas in ratio to a rate of feedstock. A difference is that transient processes in the novel reactor are ended faster in 10-100 times.

20 To remove a coke from the reactor and the apparatuses for quenching cracked gas it is necessary to stop supply of hydrocarbon feed into the preheater 1. The temperature in the working cavity of the reactor must be maintained much the same as the temperature during pyrolysis. In the result of gasification reactions a superheated steam removes the coke deposits from the reactor 4 as well as from transfer tubes and apparatuses 2 and 3 for quenching cracked gas. Outgoing flow is conveyed into afterburning installation (not shown in drawings). The process of decoking is controlled through analysis of outgoing flow. When the contents of
25 carbon oxide and carbon dioxide fall to beforehand assigned values, decoking is stopped and supply of hydrocarbon feed is renewed. The advantage of such burning-out in comparison with air or air-steam burning-outs are endothermicity of the proceeding reactions eliminating a danger of local overheating construction materials. The another advantage is that reducing ~~[conditions]~~ medium inside the reactor remains at all regimes of operation, enabling fabrication
30 of the most important reactor elements of the heat-resistant alloys on base of heat-proof metals unstable under oxidizing conditions, for example of the alloys on base of ~~[wolfram]~~ tungsten or molybdenum, or niobium.

In the present process the pyrolysis of feedstock proceeds in series in the working cavity, and further in transfer pipelines conveying pyrolyzed gas to the quenching apparatuses. The

hydrodynamic regime realized within the reactor working cavity is close to regime peculiar to apparatus of ideal mixing, where concentrations of reactants are ~~[homogenized through-out]~~ *homogeneous within* ~~[the area of]~~ this cavity. The process essentially differs by this from process realized in ~~[traditional]~~ *conventional* tubular reactors and from the process disclosed by
 5 US Patent 4265732 as well.

Since the pyrolysis in the reactor working space proceeds in presence of just cracked products of high concentration, the pyrolysis reactions are accelerated due to autocatalysis phenomenon. Because of ~~[this]~~ *it* the pyrolysis can be carried out also at lower temperature thus increasing the process selectivity. This is confirmed by experimental data on pyrolysis of
 10 hydrocarbons in presence of hot pyrolyzed gases, indicated in US Patent 3579601.

Inside transfer pipelines the pyrolysis proceeds adiabatically, without supply of heat from outside. The hydrodynamic regime realized here is close to regime peculiar to apparatus of ideal displacement as in tubular reactors. ~~[The available]~~ *This* adiabatic run enables to economize on energy *expense*~~[d]~~ for carrying out the pyrolysis process and enables also to
 15 increase the olefin yields.

The rotary blading reactor (Fig. 2 and 3) for hydrocarbon pyrolysis comprises a housing including casing 8 with lids 9 and 10. Inner surfaces of the casing 8 and the lids 9, 10 are covered with heat insulators 11, 12 and 13. The housing comprises also a heat resistance skin formed by element 14 fastened together with the casing 8, and elements 15 and 16 fastened
 20 together with the lids 9 and 10. With the element 14 the directing blades 17 and 18 are fastened together. Directing blades 19 and 20 are fastened together with the elements 15 and 16. The casing 8 is provided with nipples 21 for supplying feedstock and nipples 22 for carrying off the cracked gas. A rotor consists of shaft 23 and working wheel 24 provided with working blades 25. The latter form the blade crown of the working wheel 24. The shaft 23 is supported by
 25 radial 26 and radial-thrust 27 bearings and is sealed up by double labyrinth packings 28 and 29 into channels of which a steam is injected from outside (not shown in the drawings).

Each working blade 25 (Fig. 4 and 5) has a tail 30, by which they are fixed on a rim of the working wheel 24 forming a dovetail lock. In the blade 25 the radial channels 31 are made for passing a mixture of feedstock with steam-diluent used as cooling agent.

30 The reactor operates in the following way. Through the supplying nipples 21 a mixture of evaporated feedstock with steam-diluent enters into a gap between the casing 8 and the heat resistant skin, passes further through the channels made in the working wheel 24 and in the working blades 25 and enters into the annular working cavity where the working blades 25 and the directing blades 17, 18, 19, 20 are arranged. Processed fluid being circulated ~~[along]~~ *within*

the annular cavity comes into multiple contacts in turn with the stationary directing blades and rotating working blades, thereby creating streamlines in the form of two spirals rolled up into vortex rings of right and left directions. Thus two communicating [~~loops~~] *circuits* of pyrolyzed gas circulation are created. A heat being absorbed by endothermic reactions, which continuously proceed within the reactor working space, is compensated by influx of heat arose from converting kinetic energy into heat.

A power transferred into the said [~~circulation-loops~~] *circuits* is proportional to [~~product of~~] the rotor peripheral velocity and meridional velocity and density of the processed fluid. With increase of meridional velocity the power, dissipated by rotor, increases until this velocity reaches 0.64-0.7 sonic velocity in this fluid, e.g. about 400-500 m/s. At higher meridional velocities the power dissipated by rotor into reaction zone sharply decreases because of decrease of the fluid density averaged along a height of the blade. This is connected with a fact, that in the core of vortex ring the pressure is lower than on its periphery, and this [~~lowering~~] *pressure decrease* depends on [~~the~~] meridional velocity of the fluid.

During every passing [~~over~~] *through* [~~the-working~~] *blade crown of a working wheel* the reacting fluid acquires an additional kinetic energy, which for the time before next passing is converted into a heat. This occurs partly due to passing the fluid through stationary compression shocks arising in places of local transition of the fluid through sonic barrier, and partly due to vortex formation. The additional kinetic energy is proportional to [~~product-of~~] the rotor peripheral velocity and meridional velocity of the fluid. For example, at the rotor peripheral velocity 300-400 m/s this energy can be about 70-150 kJ/kg. During the time of residence within [~~circulation-loop~~] *the circuit* every particle of feedstock should pass over the rotor working blades several tens times on average.

The advantage of the rotary blading reactor in comparison with [~~traditional~~] *conventional* tubular reactors is that its walls [~~defining~~] *bounding* the reaction zone are not used for heat transfer, therefore, in absence of cooling *of* the walls, their temperature differs from the temperature of reacting fluid insignificantly. Lower wall temperature and pressure in the reaction zone in comparison with the same in [~~traditional~~] *conventional* tubular reactors enable to expect [~~for~~] increase in the process selectivity and increase in desired product yields when processing the same kinds of feedstock. Short time of the installation starting and possibility of full automation of its operation and control contributes to increase in the desired product yields.

The apparatus for quenching cracked gas (Fig. 6) includes "tube-in-tube" heat exchanger

comprising outer tube 32 and inner tube 33, inlet 34 and outlet 35 nipples for conveying cracked gas, inlet 36 and outlet 37 nipples for conveying cooling agent communicated with intertubular space, and ejector comprising nozzle 38, admission chamber 39 and mixing chamber 40. The nozzle 38 is connected with the inlet nipple 34. The inner tube 33 is connected by one end with the mixing chamber 40 and by another end through the tee 41 with the admission chamber 39 and the cracked gas outlet nipple 35. Outer surfaces of the quenching apparatus are covered by heat insulator 42.

Hot gas entering through the nipple 34 is formed by the nozzle 38 into a jet which sucks cooled gas being in the admission chamber 39. Resulting mixture flowing through the inner tube 33 cools down, giving back its heat to the fluid flowing through the intertubular space between the outer tube 32 and the inner tube 33 in direction from the nipple 36 to the nipple 37. Some part of cooled mixture enters into the admission chamber 39, and residual part is removed out of the apparatus through the nipple 35.

Outgoing from the nipple 35 cooled cracked gas can have the temperature about 350-400°C. Mass rate of the mixture in the inner tube exceeds about in 2-2.5 times the rate of cracked gas coming from the reactor, and the mixture being formed in the mixing chamber 40 can have the temperature about 620-660°C.

Duration of cracked gas quenching is defined only by time of mixing the jet formed by the nozzle 38 with cooled gas from the mixing chamber 40 and can be of some milliseconds. Diameter of the inner tube 33 must be enough that it should not cause considerable pressure resistance.

A heat exchanger included in the apparatus for quenching cracked gas can be of any different structure than "tube-in-tube" type. ~~[Subtle]~~ *Small* difference in pressure between heated and cooled fluids and low values of pressure enable to apply a plate heat exchanger or any others having less both a mass and a cost in comparison with quenching apparatus used usually.

Commercial applicability

The invention is intended for use in ethylene production plants as modernized as newly constructed, instead of tubular pyrolysis furnaces. Composition of pyrolysis products prepared on the novel process differs insignificantly from one in ~~[existing]~~ *existing* furnace installations operating with the same feedstock. Therefore a use of the process should not practically demand to change fractionating and gas separation plants.

For novel installations there can be used gas turbine engines of small size without intermediate preheaters and coolers of a working fluid. ~~[Efficiency of such engines is usually~~

~~26-35% at the temperature of exhaust gas 400-500°C.]~~ Best samples of such engines can have efficiency 42% at the temperature of exhaust gas 570°C. High temperature of exhaust gas enables to use effectively its heat for evaporating and preheating feedstock, for steam-diluent preparation and also for superheated steam generation in amount enough to drive compressors of a gas separation plant. As a fuel there can be used natural gas or methane-hydrogen fraction separated from cracked gas in gas separation plant. A noise made by gas-turbine engine does not exceed the noise made by acoustic gas burners in ~~[traditional]~~ *conventional* pyrolysis furnaces. To carry off an exhaust gas from gas-turbine engines there can be used chimneys similar to ones usually used in tubular furnaces. For pyrolysis plant with capacity on hydrocarbon feed about 80,000 tonnes per year it should be required a gas-turbine engine of power about 8-12 megawatts. Such engines are fabricated serially and are capable of long-time operation under conditions of continuous loading, as gas-blowers in gas pumping stations of arterial gas pipelines. Full durability of such engines can run up to 100,000 hours.

In designing novel pyrolysis installations it might be as practicable to use materials, operational experience and technologies used by producers of gas-turbine engines. If a reactor and its gas-turbine drive will be made in the same factory, the pyrolysis installation can be configured on a common framework in the form of compact aggregate of full readiness.

[Claims]

That which is claimed is:

5 ~~[1. A process for producing low molecular olefins by pyrolysis of hydrocarbons, which comprises preheating and evaporating a starting feedstock, mixing the same with a steam diluent, heating a resulting mixture to pyrolysis temperature in a blading rotary reactor by heat generated inside a volume of the mixture due to hydrodynamic drag of the rotor blades rotating therein, quenching a cracked gas and subsequent separation of it, wherein the said heating the mixture to the pyrolysis temperature is performed by mixing with hot~~
 10 ~~pyrolyzed gas being circulated in a working cavity of the blading rotary reactor for a negligible time in comparison with a duration of pyrolysis reactions].~~

~~[2. The process of claim 1, wherein the said preheating the feedstock and steam diluent is performed in two stages, and in the second stage the preheating is carried out in a heat exchanger by utilizing a heat contained in the cracked gas outgoing from the blading rotary~~
 15 ~~reactor.]~~

~~[3.] A reactor for pyrolysis of hydrocarbons in production of low-molecular olefins, comprising [a housing, with directing stationary blades,] an inlet nipple for supplying feedstock, an outlet nipple for carrying off cracked gas, [and] a working wheel [provided] with [a] blades, [crown,], stationary blades and a housing provided with a cavity, the said working~~
 20 ~~wheel and the said stationary blades are located in the said cavity and the said cavity is configured so, that hot pyrolyzed gas being in it can repeatedly passes through the same blades of the said working wheel and through the same said stationary blades, [wherein]~~

~~[the said housing has an annular cavity for circulation of hot pyrolyzed gas, which contains the directing stationary blades and surrounds the blade crown of the working wheel along~~
 25 ~~periphery], and the said inlet nipple and said outlet nipple are communicated with the said cavity.~~

~~[4. The reactor of claim 3, wherein the housing consists of a casing and a heat resistant skin fastened together, and the casing is covered on the inside by heat insulation.~~

~~5. An apparatus for quenching cracked gas comprising a heat exchanger having spaces~~
 30 ~~for cooled and heated fluids separated by a wall, wherein the said apparatus is provided with a tee and with an ejector comprising a nozzle, admission chamber and mixing chamber, so that the mixing chamber of the ejector and one of nipples of the tee are communicated with the space of cooled fluid, and the admission chamber of the ejector is connected with the another nipple of the tee.]~~

Abstract

5 A process and apparatuses for producing low-molecular olefins by pyrolysis of hydrocarbons comprising preheating and evaporating a starting feedstock, mixing the same with steam-diluent, thermal cracking a resulting mixture in a blading rotary reactor, quenching cracked gas and subsequent separation of it. The heating reacting mixture to pyrolysis temperature is performed by mixing with hot pyrolyzed gas being circulated in a working cavity of the blading rotary reactor. The process enables to increase the low-molecular olefins yield.